cm^{-1}. The predicted wave numbers tabulated by Bhatia in Table III of his paper (Ref. 10) should, therefore, be reduced by this amount.

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Zero-Field Intensity Oscillations Following Impulsive Excitation of Hydrogen*

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In a beam-foil experiment, we have observed zero-field oscillations of approximately 25% in the intensity of H_β radiation polarized parallel to the beam. An unmodulated decay is observed when a signal proportional to the total intensity is examined. Our observations substantiate Macek's theory of coherent decay from fine-structure levels.

The first evidence that the decay of radiation from an excited state might show an oscillatory time dependence under zero-field conditions was provided in an experiment by Bashkin and Beauchemin.[1] They observed the decay of helium ions impulsively excited in a carbon foil. Macek[2] suggested that such oscillations could be caused by an interference between fine-structure levels, under completely field-free conditions. Subsequently, Andrä[3] observed zero-field oscillations in helium and for $n=3, 4$ in hydrogen. In the case of H_β the data could be decomposed into frequencies 1390 and 440 MHz and possibly 1500 MHz. These frequencies represent the fine-structure separations of $p_{1/2}$-$p_{3/2}$ (1371 MHz, $d_{3/2}$-$d_{5/2}$ (457 MHz), and a possible mixture of $s_{1/2}$-$d_{3/2}$ (1233 MHz and $s_{1/2}$-$d_{5/2}$ (1690 MHz). Sellin, Biggerstaff, and Griffin[4] reported on a similar experiment on H_β and H_γ, and essentially observed no oscillations within the limits set by their apparatus. Andrä's work was performed in the earth's magnetic field where the motional electric field experienced by the moving atom ($\vec{v}\times\vec{B}$) is approximately 2 V/cm. Sellin, Biggerstaff, and Griffin performed their experiment in a chamber which was surrounded by Helmholz coils which reduced the magnetic field to 50 mG. We have successfully tested two facets of Macek's interpretation of zero-field oscillations.

All present observations were made with carbon foils of nominal thickness 10 $\mu g/cm^2$.[5] The 130-keV proton beam was collimated by two $\frac{1}{8}$-in. apertures, the first being located in the beam pipe and the second directly in front of the foil. These apertures were separated by an initial distance of 13 cm (see Fig. 1). Subsequent to excitation, observations were made on the 4861 Å H_β emission from the hydrogen component of the beam, whose velocity was 5×10^8 cm/sec.[6] Beam currents of 15–20 μA were collected in the Faraday cup.

The foil and decay region were enclosed within a magnetic shield. The component of the magnetic field normal to the beam was measured to be 37.6 mG. The motional electric field experienced by an atom in the beam was thus 0.19 V/cm.

The radiation was detected by a 1-m, normal incidence, McPherson Model No. 225 monochromator with a reciprocal dispersion of 16 Å/mm, using 1-mm slits and an uncooled EMI-6256S photomultiplier. The signal was processed by a conventional counting system (Fig. 1), with the scaler gated from a current integrator set to stop the scaler when a preset charge was accumulated in the Faraday cup (usually 12×10^{-4} C). A rotatable polarizer (type HN22 Polaroid) was placed close to the entrance slit of the mono-

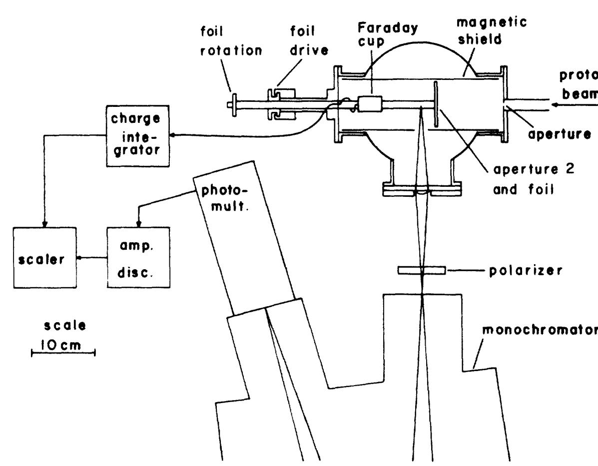

FIG. 1. Foil drive mechanism and optical detection system.

chromator.

Following Macek,[7] an atom which is impulsively excited at $t=0$ is described by the wave function

$$\psi(t)=\sum_{JM} a_{JM} U_{JM} \exp\left[-\left(\frac{iE_J}{\hbar}+\frac{\gamma_J}{2}t\right)\right], \tag{1}$$

where a_{JM} is the amplitude for exciting the state JM, U_{JM} is an eigenfunction of the time-independent Schrödinger equation, E_J is the energy of excitation of the JM state, and γ_J is the decay constant for state JM. The photon flux (henceforth referred to as intensity) associated with decay to a lower state J_0M_0 is given by

$$I_q=\sum_{M_0}|\langle\psi(t)|V_q|J_0M_0\rangle|^2, \tag{2}$$

where V_q is the qth component of the dipole length or dipole velocity operator. It is the existence of cross terms in the expansion of the square of the matrix element which lead to oscillations. For the case of H_β we consider initial states $S_{1/2}$, $P_{1/2}$, $P_{3/2}$, $D_{3/2}$, and $D_{5/2}$. We now expect to observe a decay of the form

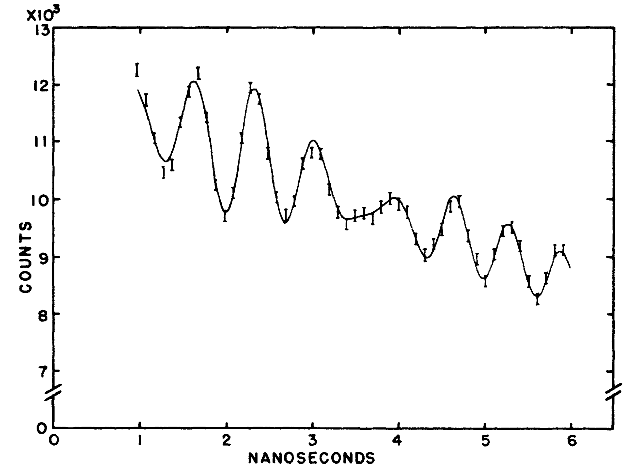

FIG. 2. Decay pattern for radiation polarized parallel to the beam. Error bars indicate a statistical uncertainty of ±1 standard deviation. Solid line is a computer fit to the data.

$$I_q=A_1\exp(-\gamma_s t)+A_2\exp(-\gamma_p t)(A_3+\cos\omega_{p_{1/2}p_{3/2}}t)+A_4\exp(-\gamma_d t)(A_5+\cos\omega_{d_{3/2}d_{5/2}}t)$$
$$+A_6\exp[\tfrac{1}{2}(\gamma_s+\gamma_d)t][0.4\cos(\omega_{sd_{3/2}}t)+0.6\cos(\omega_{sd_{5/2}}t)], \tag{3}$$

where γ_s, γ_p, and γ_d refer, respectively, to the decay constants of the S, P, and D states, $\omega_{p_{1/2}p_{3/2}}$ is the frequency separation, $(E_{3/2}-E_{1/2})/\hbar$, between $P_{1/2}$ and $P_{3/2}$, etc. The constants A_1-A_6 depend upon excitation amplitudes, transition probabilities, and Clebsch-Gordan coefficients. The coefficients of the s-d coherence cosine terms are determined by the squares of the appropriate Clebsch-Gordan coefficients and have been evaluated by Macek[8] in an extension of Table I, Ref. 6, to be 0.4 for s-$d_{3/2}$ and 0.6 for s-$d_{5/2}$.

Figure 2 shows the decay pattern we have observed for H_β radiation polarized parallel to the beam, with time $t=0$ defined at the position of the foil. (The length of the beam sampled at each data point is 0.85 mm.) No data points are shown close to the foil where the light path to the lens is interrupted by the foil holder mechanism.

We have used the expression (3) above with six variable coefficients A_1–A_6 and additionally three phase angles to obtain a fit to the experimental data. This was done with a computer program which uses a search procedure to minimize the χ^2 of the data to the trial fitting function integrated over the slit function of the monochromator. The best fit to the data was given by the function

$$I_q=1466e^{-t/230}+744e^{-t/12.4}[11.9+\cos(8.614t+5.02)]+98e^{-t/36.5}[21.0+\cos(2.871t+5.03)]$$
$$+765e^{-t/133.2}[0.4\cos(7.747t+0.51)+0.6\cos(10.618t+0.51)], \tag{4}$$

where t is the time in nanoseconds. Lifetimes used are obtained from the work of Bethe and Salpeter.[9] The solid line in Fig. 2 represents this function integrated over the slit function. For this run the χ^2 per point is 1.03. Without use of the phase angles, which take account of phase differences for the initial wave functions, a χ^2 of 22 per point was the best fit obtained.

Our results indicate that the interference between the $d_{3/2}$ and $d_{5/2}$ states, involving the relatively slow component at 457 MHz, makes a small contribution to the oscillatory part of the decay. However, the sd coherence terms between levels of different angular momentum (suspected by Andrä in his analysis of the H_β emission) are strongly present and their existence is firmly established. The closeness of the functional fit to the experimental data justifies Macek's interpretation of zero-field beats.

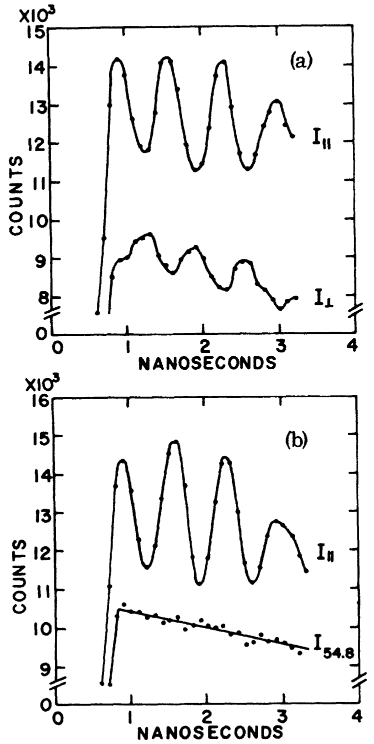

FIG. 3. (a) Comparison of decay patterns for radiation polarized parallel and perpendicular to the beam. Relative detection efficiency of the monochromator: $k_{\parallel}/k_{\perp}=1.31$. (b) The oscillations are removed from the decay pattern when the polarizer is placed at 54.8°. A different foil than that in (a) was used in this run. All solid lines are hand drawn curves through the data points.

In our second test of Macek's theory we have investigated the prediction that the total intensity radiated by the beam will be unmodulated. We have examined the intensity radiated with electric vector parallel and then perpendicular to the beam. This is shown in Fig. 3. The patterns, shown in detail for approximately 3 nsec, are reversed in phase. This is as one should expect if the total intensity is unmodulated. We have used the polarizer at a "magic" angle to observe an intensity proportional to the total intensity.

The total intensity radiated by the beam is

$$I_t=(8\pi/3)(I_{\parallel}+2I_{\perp}), \tag{5}$$

where $I_{\parallel}$ and $I_{\perp}$ represent intensities radiated at 90° with respect to the beam and whose polarization axes are, respectively, parallel and perpendicular to the beam. Suppose we now place a polarizer with its polarizing axis at an angle θ with respect to the beam direction. Let $k_{\parallel}$ and $k_{\perp}$ be the relative detection efficiencies of the optical detection system to light with electric vectors parallel and perpendicular to the beam. Then the intensity transmitted by the polarizer is

$$I_{\parallel}\cos^2\theta+I_{\perp}\sin^2\theta \tag{6}$$

and the light is now polarized at an angle θ with respect to the beam. Thus the detection efficiency of the monochromator/photomultiplier will be $k_{\parallel}\cos^2\theta+k_{\perp}\sin^2\theta$ and the total transmitted intensity for the polarizer plus detector will be[10]

$$(I_{\parallel}\cos^2\theta+I_{\perp}\sin^2\theta)(k_{\parallel}\cos^2\theta+k_{\perp}\sin^2\theta). \tag{7}$$

If we choose $\sin^2\theta=\frac{2}{3}$, then $\cos^2\theta=\frac{1}{3}$ and we combine $I_{\parallel}$ and $I_{\perp}$ in the ratio 1:2. For this particular "magic" angle, 54.8°, $k_{\parallel}\cos^2\theta+k_{\perp}\sin^2\theta$ is a constant, effectively the efficiency of the system to light polarized at 54.8°. Hence the signal measured by the detection system will be proportional to the total intensity. Figure 3 shows the result obtained when the polarizer is set at 54.8°. The oscillations are effectively cancelled and we obtain a decay containing no oscillations, again in full agreement with Macek's prediction.

In addition we would like to point out that this technique has important applications in beam-foil spectroscopy. In general one expects to observe oscillations associated with the coherent decay of excited states, even when the detection optics do not selectively detect light of different polarizations. (This follows because the oscillatory component of the parallel polarized radiation is a factor of 2 larger than the equivalent perpendicular polarization.) In some cases this will lead to a scatter in the experimental points about some decay which is expected to be exponential. This problem can be removed entirely by using a fixed polarizer at 54.8°. The signal will be decreased (in our case by a factor of 4) but the scatter will be significantly reduced. In addition it would be advisable to shield the decay region from stray magnetic and electric fields, which may otherwise lead to Stark splittings causing oscillations in the observed decay. However for Stark perturbations the total intensity is modulated and the polarizer will not remove the oscillations.

We are most grateful to our colleague Dr. Joseph Macek for many enlightening and stimulating discussions.

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Measurement of the g Factor of Conduction Electrons by Optical Detection of Spin Resonance in p-Type Semiconductors

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A new optical method to detect conduction-electron spin resonance in semiconductors is described. Its main interest is its application to undoped samples in which the properties of free carriers are not perturbed by shallow donor impurities. The g factor of 10^8 photocreated electrons at the bottom of the conduction band in p-type GaSb was measured to be $|g^*| = 9.3 \pm 0.3$. It is different from most of the previous experimental determinations and from the theoretical prediction $g_{th}^* = -6.66$.

An optical detection technique has been used for the first time to detect conduction-electron spin resonance (CESR) in semiconductors. Free electrons were produced at the bottom of the conduction band by light irradiation in p-type GaSb at liquid-helium temperature. We observed the resonance line of the photocreated electrons by analyzing the polarization of the recombination light. This yielded the only resonant determination of the g factor of conduction electrons in this material. The measured value $|g^*| = 9.3 \pm 0.3$ is significantly greater than the predicted one, $g_{th}^* = -6.66$.[1] It is also greater than the g value obtained from various experiments in high magnetic fields.[2-6] The resonance linewidth of the photocreated electrons is due partly to the finite lifetime τ of the free carriers, and due partly to the spin relaxation time T_1 in the conduction band. The half-width is $\Delta H = 3.50 \pm 0.25$ G at 1.7°K in the applied external magnetic field $H \sim 12$ G.

Optical detection techniques of spin resonance, together with optical pumping,[7] have been extensively used in gases to measure Landé factors, hyperfine constants, and relaxation parameters.[8] In solids, these methods have been applied to the study of localized paramagnetic impurities.[9] Optical pumping on delocalized states in semiconductors[10] and optical detection of the conduction-electron polarization[11] have already been achieved in a few cases. We have applied a resonant excitation to the photocreated electronic spins and have obtained directly the g factor at the bottom of the conduction band.

The principle of optical detection of CESR is the following: In a semiconductor, the recombination light is circularly polarized whenever the free-electron gas is spin polarized. This effect has been observed in GaSb[11] and $Ga_{1-x}Al_xAs$[12] where the electronic polarization was produced by optical pumping. The spin-polarized electrons are placed in a dc longitudinal magnetic field H, parallel to the direction of spin polarization. Transitions between the two spin levels are induced by a transverse rf magnetic field at frequency $\omega/2\pi$ when the resonance condition $\hbar\omega = g^*\mu_B H$ is fulfilled (μ_B is the Bohr magneton). These transitions produce a change ΔP in the steady-state electronic polarization P, and the polarization of the recombination light is affected. Spin resonance is thus detected by a change in the optical signal.

The signal, being proportional to the change ΔP of the spin polarization, is proportional to the degree of saturation of the spins. To obtain an appreciable saturation one needs a transverse rf field of the order of the linewidth ΔH of the resonance line. This is more conveniently achieved at a low frequency which corresponds to a low magnetic field H. A large electronic polarization P is obtained in the field of the experiment, $H \sim 12$ G, by optical pumping with cir-